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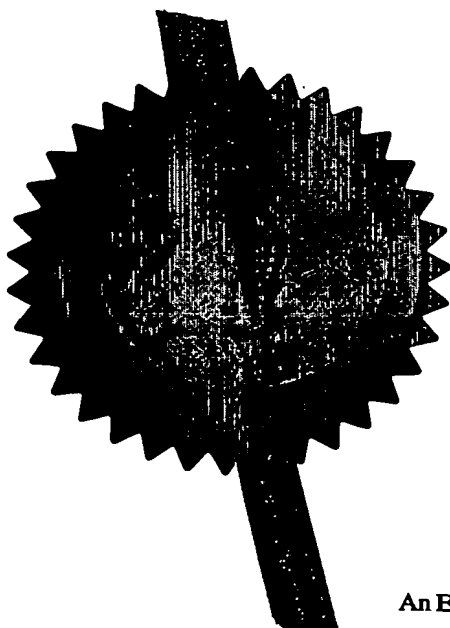
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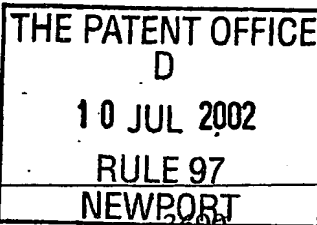
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1. Your reference

SMR/MAW/P550679

2. Patent application

(The)

0216052.1

10 JUL 2002

11JUL02 E732530-1 D00637

3. Full name, address and postcode of the or of each applicant (underline all surnames)

Arjobex Limited
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Patents ADP number (if you know it)

If the applicant is a corporate body, give the country/state of its incorporation

United Kingdom

6459655003

4. Title of the invention

Synthetic Paper Label

5. Name of your agent (if you have one)

"Address for service" in the United Kingdom to which all correspondence should be sent (including the postcode)

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Patents ADP number (if you know it)

4332719001

6459655003

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UK 1/77
9/5/03

Country	Priority application number (if you know it)	Date of filing (day / month / year)
UK	1/77	9/5/03

7. If this application is divided or otherwise derived from an earlier UK application, give the number and the filing date of the earlier application

Number of earlier application

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- a) any applicant named in part 3 is not an inventor, or
 - b) there is an inventor who is not named as an applicant, or
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Description

Claim(s)

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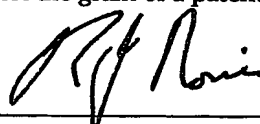
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Any other documents (please specify)

11. I/We request the grant of a patent on the basis of this application.

Signature



Date

10 July 2002

12. Name and daytime telephone number of person to contact in the United Kingdom

R.J. NORRIS Appointed Agent

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SYNTHETIC PAPER LABEL

- 5 The present invention relates to a composite label made from plastics film or sheet having an external printable surface, for example a synthetic paper.

The term "synthetic paper" is used herein and throughout the specification to mean plastics film and sheet products having a feel and printability similar to cellulose paper.

- 10 It has been recognised that plastics sheet material of this type can provide an improved alternative to paper where durability and toughness are required. Synthetic papers produced from polyolefins have several advantages over other plastics since they offer UV resistance, good tear strength and the ability to be recycled in many post-consumer waste applications. Labels are amongst the established uses of such products.

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In certain labelling applications, it is desirable that the label construction should be such as to reveal any attempt at tampering after it has been applied, i.e. it should be "tamper evident". This is particularly important if the label is to be applied to high value products such as cosmetics, perfumes and fine wines and spirits.

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In order to be fully tamper-evident, the label should exhibit internal cohesive failure if attempts are made to peel the label off the surface to which it has been applied. Preferably, it should also shear through its full thickness, because delamination without accompanying shearing through the full thickness of the product could leave the printed surface of the label intact and therefore available for fraudulent purposes. However, a label which delaminates but does not shear as just described would nevertheless represent worthwhile progress in the art.

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- 30 It is an object of the invention to provide a synthetic paper or similar tamper-evident labelling material which meets the first, and preferably also the second, of the above-stated requirements.

Accordingly, the present invention provides, in a first aspect, tamper-evident labelling material comprising synthetic paper or other composite plastics sheet material which is

printable on its upper surface, and comprises a base layer, typically of polyolefinic material, and optionally a printable layer forming the printable surface, characterised in that the base layer has a density of not more than 0.6 g cm^{-3} , preferably not more than about 0.5 g cm^{-3} .

In a second aspect, the invention resides in the use, in synthetic paper or other printable composite plastics sheet labelling material for the purpose of imparting tamper-evident properties, of a sheet material which comprises a base layer, typically of polyolefinic material, and optionally a printable layer forming the printable surface, characterised in that the base layer has a density of not more than 0.6 g cm^{-3} , preferably not more than about 0.5 g cm^{-3} .

Preferably, the labelling material comprises a co-extruded core layer between upper and lower skin layers, all typically of polyolefinic material, and optionally a printable coating on the upper skin layer, wherein the core layer has a density of not more than 0.6 g cm^{-3} , preferably not more than about 0.5 g cm^{-3} , and the skin layers have a density of more than 0.6 g cm^{-3} , preferably more than 0.9 g cm^{-3} . The skin layers preferably have a thickness of not more than $2\mu\text{m}$.

The mean cohesive strength of the labelling material, as measured by the tape pull test described below, is preferably not more than about 3N (Newtons) if shearing through the full thickness of the material is to occur when an attempt is made to peel off the label.

The tape pull test involves measuring the force required to peel off an adhesive tape adhered to the labelling material. The effect of the removal of the tape on the integrity of the labelling material is also observed. The test may conveniently be carried out using 25mm wide transparent tape of the kind supplied for domestic and office use, for example that sold as "Sellotape* Original" (here and elsewhere in this specification, an asterisk indicates a proprietary trade mark.). The tape is pressed into contact with the surface of the labelling material by hand (normally in the machine direction, if this is known) and the resulting bond is reinforced by running a 1kg roller over the tape. The tape is then peeled away from the labelling material using a so-called "universal tester" (for example as supplied by Instron Limited, United Kingdom) at a defined speed

(typically 250 or 500 mm min⁻¹) and defined contained angle (180°). The average force required for this over the duration of the peeling operation is recorded, and provides a measure of the cohesive strength of the material under test. As already mentioned, average values for this peeling force should be less than 3N. There should be clear delamination and breakaway of the label material; in other words the label should break in two through the thickness of the film after an initial delamination step.

Low-density synthetic paper or other composite plastics sheet material as described above is easily converted into pressure-sensitive adhesive labels, unlike tamper-evident sheet materials produced from PVC or polystyrene, which tend to be brittle and difficult to handle.

The synthetic paper or other composite plastics sheet material used in the present invention can be produced by generally known techniques, for example simultaneous biaxial orientation techniques as disclosed in British Patents Nos. 1374574 and 1442113 in combination with voiding and/or co-extrusion techniques such as disclosed in British Patents Nos. 1470 372, 1490512 or 1492771, European Patent No. 470760B or European Patent Application No. 863177A. The proportion of voids must be such as to produce the desired low base layer density and internal cohesive strength. Fillers, rubbers, antioxidants, lubricants, antistatic agents and other additives may be included in the sheet material, as is conventional in the synthetic paper art.

Preferably, a synthetic paper comprising high density polyethylene (HDPE) is used. The term high density polyethylene as used herein refers to polymers and copolymers of ethylene with minor amounts of other α -olefin monomers as is commonly understood in the industry and as described in the above-mentioned patent publications. The term is also used to refer to mixtures of high density polyethylenes.

Plastics substrates are generally impervious to materials such as printing inks. Thus, they tend to be unsuitable for direct use in ink-based printing processes because the ink has a tendency to remain at the surface of the substrate, without being absorbed. It is well known to modify the surface characteristics of a plastics substrate by applying an absorbent coating composition to the surface of the substrate. Such a coating

composition comprises an absorbent filler, which renders the substrate more absorbent to ink, and a polymeric binder, which binds the filler to the surface of the substrate. Prior to application of the coating composition, the surface of the substrate is subjected to flame or corona discharge treatment such that the treated surface has a surface energy of about 47 dynes cm^{-1} . Alternatively, if no separate printable coating is to be applied, and the upper skin layer is to provide the printable surface, such a surface treatment can be carried out to provide a more polar surface and the required wettability to more readily accept inks and coatings. The surface may also be suitably textured or modified by the presence of a pigment and/or a filler, and suitably has a printability corresponding to a wettability of at least 40 dynes cm^{-1} .

When a printable surface coating is used, it may be formulated in a similar manner to aqueous coatings as commonly used in the paper industry and especially latex-based coatings. Of particular value in the present context, however, are the coatings described in British Patent Application No. 2177413A and European Patent Application No. 1055711A.

Preferably such a printable coating is provided by the application of an aqueous system comprising a polymeric binder, an absorbent pigment and an antistatic agent. Typically the binder: pigment dry weight ratio is in the range 15:100 to 50:100, preferably 22:100 to 35:100, and the antistatic agent : pigment dry weight ratio is from 0.4:100 to 2.5:100. The antistatic agent may be, for example, sodium polyphosphate or another of the agents disclosed in British Patent Application No. 2177413A.

The polymeric binder may be in an aqueous or latex suspension, preferably a latex suspension, and should contain carboxyl groups on the polymer chain of at least one polymeric constituent. The binder may comprise a single polymer or a mixture of polymers. The binder may comprise, for example, starch or protein modified chemically or by physical addition of other polymeric species. Alternatively the binder may comprise a carboxylated styrene-butadiene copolymer, an acrylic polymer or copolymer, or a vinyl acetate polymer or copolymer. Preferably, the binder comprises a carboxylated styrene-butadiene copolymer.

The binder content of the aqueous coating composition may be chosen to suit individual requirements, for example, the stiffness of the coated product. Preferably, the binder content of the composition is in the range 15 to 30% by weight based on the binder plus aqueous phase, and more preferably in the range 20 to 25%.

The absorbent filler may be any dispersible solid but is preferably an inorganic filler or pigment such as calcium carbonate, china clay, or titanium dioxide.

- 10 The composition may also contain an insolubilizing agent, preferably a polyanionic zirconium compound, most preferably ammonium zirconium carbonate. If desired, security inclusions such as fluorescent pigments and/or coloured fibres can also be present.
- 15 The labelling material can be coated with adhesive on its lower surface and converted into labels in conventional manner.

The invention will now be illustrated by the following Example, in which all parts and percentages are by weight unless otherwise specified, and all references to melt flow index (MFI) values are as measured by ASTM Method No. 1238 :

A synthetic paper comprising an extruded core layer and two upper and lower co-extruded skin layers was first produced.

- 25 The core layer was derived from a compound (Mix A) consisting of a mixture of HDPE having an MFI of 0.33; particulate filler (CaCO_3 , TiO_2); voiding agent (a pentaerythritol - maleic anhydride derivative of abietic acid) and processing aids, as follows:

Mix A

Component	Wt%
Ampacet* 111096 60% CaCO ₃ in HDPE	15.9
ExxonMobil HDPE Paxon* AL55-003	55.9
Ampacet* 111096 60% TiO ₂ in HDPE	8.00
Nova Chemicals Polystyrene High Heat 1300	3.95
Omya Omyalene* G.200 88% CaCO ₃ in LDPE	8.00
DRT Dertoline* DP 001 voiding agent	8.00
Ciba Geigy Irganox* B215 anti-oxidant	0.22

The two co-extruded outer layers were each derived from a dry blend mixture of HDPE having an MFI of 0.33 and linear low density polyethylene (LLDPE) having an MFI of 2.0, as follows:

Mix B

	wt %
ExxonMobil HDPE Paxon* AL55-003	75
ExxonMobil LLDPE LL-1002	25

Mix A was compounded at 210 °C by a conventional extrusion, dicing and cooling process. This compound was introduced into the primary extruder of an in-line twin extruder – distributor – sheeting die coextrusion arrangement. Mix B was introduced into a coextruder feeding the distributor. The sheeting die and distributor were of a conventional type enabling a three layer coextrudate to be produced continually, comprising a homogeneous layer of Mix B on each side of a layer of Mix A. The throughput of the in line primary/secondary extruders was arranged to be 650 kg per hour from screw speeds of 22 and 33rpm respectively, with an exit temperature of 191° C. The throughput of the coextruder was 28 kg per hour from a screw speed of 12 rpm with an exit temperature of 185° C. The output of the extruders was fed to the distribution block held at 210° C. The die lips were adjusted to 5 mm and the sheeting die gave an extrudate of about 420 mm wide. This composite sheet was then fed directly onto and around a set of cooling and conditioning rollers having the following temperatures

ROLL 1	° C	54.0
ROLL 2	° C	76.0
ROLL 3	° C	77.0
ROLL 4	° C	90.0
COND. ROLL 5	° C	118.0
COND. ROLL 6	° C	116.0
COND. ROLL 7	° C	118.0
COND. ROLL 8	° C	121.0
COND. ROLL 9	° C	121.0
COND. ROLL 10	° C	121.0

The final conditioned composite sheet, which had an overall thickness of about 1000µm with a core thickness of 960µm and outer skin thicknesses of 20µm, was then fed into a simultaneous biaxial stretching apparatus of the type described and illustrated in British Patent No.1442113. The apparatus was arranged to produce a 4:1 stretch in each of the longitudinal or machine direction (MD) and transverse direction (TD). The stretching apparatus was provided with a 6-zoned oven: zones 1 and 2 provided preheat, zones 3 and 4 the stretching section and zones 5 and 6 the annealing section. The temperatures are given in the following table:

ZONE 1	° C	135.0±2
ZONE 2	° C	125.0±2
ZONE 3	° C	117.0±2
ZONE 4	° C	117.0±2
ZONE 5	° C	130.0
ZONE 6	° C	130.0

The composite plastics film thus produced was cooled, edge trimmed and passed through a corona discharge treatment unit such that the product had a surface energy of about 47 dynes cm⁻¹. Finally it was wound into reels approximately 1500 mm wide.

Typical properties measured for samples of synthetic paper film produced as described above are given in the following table:

	<u>MD</u>	<u>CD</u>
Tensile (N/mm ²)	39	30
Elongation (%)	100	79
Tear (N/mm)	50	76
Opacity	96	
Stiffness(Gurley)	64	60
CoreThickness (μm)	100	
Core density (g cm ⁻³)	0.50	

A printability coating was later applied to both surfaces of the film using an air-knife coater, and then dried. The approximate formulation of the coating was as follows:

5

Parts by weight

Water	100
Acrylate dispersant	5
Ground calcium carbonate of which 90% of the	
10 particles are below 2μm particle size	100
Anionic wetting agent	2
Mixture of acrylonitrile acrylate styrene copolymer	
(50% solids content) and styrene/n-butanol copolymer	
(50% solids content) in the ratio 1:4	63
15 Ammonium zirconium carbonate	4

As already mentioned, the cohesive strength of the synthetic paper film (which provides a valuable predictor of tamper-evident character) is particularly important in this application.

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Typical measured peeling force (cohesive strength) values for samples of 50 g m⁻² synthetic paper film produced as described above and having 4 g m⁻² printability coatings on each surface (i.e. total 58 g m⁻²) are given in the following table (the measurement method was as described earlier in this specification, with a peel speed of 25 500 mm min⁻¹). The maximum measured peeling force (cohesive strength) during the peeling operation was also noted.

Peel Speed Mm/min	Maximum Cohesive Strength (N)	Average Cohesive Strength (N)
250	3.15±0.15	2.2±0.15
500	3.5±0.2	2.05±0.1

5 It was observed that each sample broke away through the thickness of the substrate during the peeling operation. As explained earlier in this specification, achievement of this property is very significant in terms of a fully satisfactory tamper-evident effect.

10 When samples of synthetic paper film produced as described above were coated with a pressure sensitive adhesive laminated with release paper and die cut, the resulting labels could be easily adhered to glass containers but attempts at removal always left part of the label on the bottle, making fraudulent label transfer impossible.

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